

CLAIMS

1. A negative electrode for lithium secondary batteries, comprising a negative active material and a binder,
5 wherein the negative active material comprises graphite A and graphite B,
shapes of primary particles of the graphite A are spherical or elliptical,
an average particle diameter of the primary particles of the graphite
10 A ranges between 10 μm and 30 μm inclusive,
sizes of crystallites of the graphite A in a direction of a c-axis are smaller than 100 nm and tap density of the graphite A is 1.0 g/cm³ or higher,
shapes of primary particles of the graphite B are flat,
an average particle diameter of the primary particles of the graphite
15 B ranges between 1 μm and 10 μm inclusive, and
sizes of crystallites of the graphite B in a direction of a c-axis are 100 nm or larger.
2. The negative electrode for lithium secondary batteries according to
20 Claim 1, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.
3. The negative electrode for lithium secondary batteries according to Claim 1,
25 wherein, I_{1350} denotes Raman intensity at approximately 1350cm⁻¹, I_{1580} denotes Raman intensity at approximately 1580cm⁻¹ and a R-value of Raman spectrum is obtained by a formula: $R=(I_{1350}/I_{1580})$,
a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

4. The negative electrode for lithium secondary batteries according to Claim 1, wherein the primary particles of the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between 10 μm and 30 μm inclusive.

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5. The negative electrode for lithium secondary batteries according to Claim 1, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

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6. The negative electrode for lithium secondary batteries according to Claim 1, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.

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7. A method for manufacturing a negative electrode for lithium secondary batteries comprising the steps of:

preparing graphite A of which shapes of primary particles are spherical or elliptical, an average particle diameter of the primary particles ranges between 10 μm and 30 μm inclusive, sizes of crystallites in a direction of a c-axis are smaller than 100 nm, and tap density is 1.0 g/cm³ or higher;

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preparing graphite B of which shapes of primary particles are flat, an average particle diameter of the primary particles ranges between 1 μm and 10 μm inclusive, and sizes of crystallites in a direction of a c-axis are 100 nm or larger;

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preparing paint by mixing the graphite A and the graphite B in the presence of a binder and a solvent; and

applying the paint on a collector, drying the paint and then performing a pressure forming treatment.

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8. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.

5 9. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7,

wherein, I_{1350} denotes Raman intensity at approximately 1350cm^{-1} , I_{1580} denotes Raman intensity at approximately 1580cm^{-1} and a R-value of Raman spectrum is obtained by a formula: $R=(I_{1350}/I_{1580})$,

10 a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of 5145 \AA .

10. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein the primary particles of
15 the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between $10\text{ }\mu\text{m}$ and $30\text{ }\mu\text{m}$ inclusive.

11. The method for manufacturing the negative electrode for lithium
20 secondary batteries according to Claim 7, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

12. The method for manufacturing the negative electrode for lithium
25 secondary batteries according to Claim 7, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.

13. A lithium secondary battery, comprising a positive electrode, a negative electrode and nonaqueous electrolyte,

30 wherein the negative electrode comprises a negative active material

and a binder,

the negative active material comprises graphite A and graphite B,

shapes of primary particles of the graphite A are spherical or elliptical,

5 an average particle diameter of the primary particles of the graphite A ranges between 10 μm and 30 μm inclusive,

sizes of crystallites of the graphite A in a direction of a c-axis are smaller than 100 nm and tap density of the graphite A is 1.0 g/cm³ or higher, shapes of primary particles of the graphite B are flat,

10 an average particle diameter of the primary particles of the graphite B ranges between 1 μm and 10 μm inclusive, and

sizes of crystallites of the graphite B in a direction of a c-axis are 100 nm or larger.

15 14. The lithium secondary battery according to Claim 13, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.

15. The lithium secondary battery according to Claim 13,
20 wherein, I_{1350} denotes Raman intensity at approximately 1350cm⁻¹, I_{1580} denotes Raman intensity at approximately 1580cm⁻¹ and a R-value of Raman spectrum is obtained by a formula: $R=(I_{1350}/I_{1580})$,

a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

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16. The lithium secondary battery according to Claim 13, wherein the primary particles of the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between 10 μm and 30 μm inclusive.

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17. The lithium secondary battery according to Claim 13, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.
- 5 18. The lithium secondary battery according to Claim 13, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.